

Research



Cite this article: Abu-Bakr AF, Zubarev AYU.
2020 On the theory of magnetic hyperthermia:
clusterization of nanoparticles. *Phil. Trans. R.
Soc. A* **378**: 20190251.
<http://dx.doi.org/10.1098/rsta.2019.0251>

Accepted: 23 September 2019

One contribution of 18 to a theme issue
'Patterns in soft and biological matters'.

Subject Areas:

nanotechnology, mathematical modelling,
mathematical physics

Keywords:

bulk cluster, chain-like two particles, chaotic
orientation of easy magnetization axes,
mathematical modelling, hyperthermia

Author for correspondence:

Ali F. Abu-Bakr
e-mail: alibakrm@yahoo.com

On the theory of magnetic hyperthermia: clusterization of nanoparticles

Ali F. Abu-Bakr^{1,2} and Andrey Yu. Zubarev^{2,3}

¹Mathematics and Computer Science Department, Faculty of
Science, Menoufia University, 32511 Shebin El-Kom, Egypt

²Theoretical and Mathematical Physics Department, Institute of
Natural Sciences and Mathematics, Ural Federal University, Lenin
Ave. 51, Ekaterinburg, 620083, Russia

³M.N. Mikheev Institute of Metal Physics, Ural Branch of the Russian
Academy of Sciences, Ekaterinburg, 620108, Russia

AFA-B, 0000-0002-0444-1159; AYUZ, 0000-0001-5826-9852

Experiments show that clusters consisting of nano-sized ferromagnetic particles strongly affect the intensity of heat production during magnetic hyperthermia. In this paper, a theoretical study and mathematical modelling of the heat production by clusters of single-domain ferromagnetic particles, immobilized in a host medium, are presented. Two situations of strong and weak magnetic anisotropy of the particles are considered. Our results show that, in the case of strong anisotropy, the clusterization weakens the thermal effect, whereas in the case of weak anisotropy it enhances it.

This article is part of the theme issue 'Patterns in soft and biological matters'.

1. Introduction

Magnetic hyperthermia (MH) is a prospective and efficient method of therapy for cancer and other diseases [1–9]. The main idea of this method lies in the injection of a ferrofluid with nano-sized ferromagnetic particles into the tumour area. The particles are heated by an external alternating magnetic field. Many investigations illustrate that MH allows heating *in vivo* of the tumour region up to temperatures of 42–46°C [10]. In this temperature range, the tumour cells die, whereas the healthy cells survive. MH is especially efficient in combination with chemotherapy and radiotherapy [11]. Usually, iron oxide (maghemite, magnetite) particles

are used in the biomedical applications of MH. First, because they are not toxic; secondly, they are relatively cheap; and, next, they have sufficient magnetic moment to be detectable in the magnetic fields that are easily achievable in clinical and laboratory conditions. It was concluded [10,12,13] that particles with diameter in the range 20–25 nm and more are quite efficient for heat production. On the other hand, experiments [14] demonstrate that clusterization of the particles with diameter of about 10 nm rather enhances the thermal effect.

Theoretical methods for the quantitative prediction of heat production are required for the accurate application of the MH method for tumour therapy. The majority of the known theoretical models deal with the approximation of single non-interacting particles (e.g. [15–18]). However, the energy of magnetic interaction between iron oxide particles with diameter of about 20 nm is significantly more than the thermal energy $k_B T$. Under magnetic and colloidal interaction the particles can form aggregates of various shapes and topologies. Electron microscopic images of some of these aggregates can be found in [19].

There are two main mechanisms for the heat production by magnetic nanoparticles (e.g. discussion in [15]). The first one is rotation of the particles and energy dissipation due to the viscous friction between the particles and the surrounding medium. The second one is internal remagnetization of the particles, which takes place even when they are motionless (Néel remagnetization). Experiments show that magnetic nanoparticles embedded into biological tissues, as a rule, are tightly bound with the medium and, therefore, they are rather immobilized [2]. Therefore, the Néel mechanism must be dominating for the particles in biological tissues.

The aim of the present work is the theoretical study of the effect of particle clusterization on the intensity of heat production during MH, induced by an alternating linearly polarized magnetic field. It is supposed that the ferromagnetic particles are immobilized in the carrier environment. The limiting cases of strong and weak magnetic anisotropy of the particles are considered.

The structure of the paper is the following. In §2, the theoretical approach and the mathematical modelling of MH heat production in clusters of single-domain particles are suggested. Sections 3 and 4 introduce the mathematical models of clusters consisting of two and three particles with strong anisotropy and weak internal magnetic anisotropy of the particles. The general conclusion is presented in §5.

2. Theoretical approach and mathematical modelling

(a) Dynamics of particle magnetization

It was mentioned above that magnetic nanoparticles, being injected into biological tissue (a tumour, for instance), can form heterogeneous structures (aggregates) of various shapes and topologies. In order to get results in a physically transparent form, we consider single particles, a pair of particles and particles united in dense triangles, as illustrated in figure 1. Note that magnetic interaction between the particles in the clusters must lead to the energetically most favourable orientations of the particle axes of easy magnetization. However, in real systems, aggregation can take place also due to a not completely screened central colloidal interaction. In this case, the occasional orientation of these axes can be frozen for a very long time. That is why we suppose an arbitrary orientation of the axes in the clusters.

In a first approximation, the dynamics of the particle remagnetization can be described by using the semi-phenomenological Debye equation (e.g. [12,15]):

$$\tau \frac{dM_i(t)}{dt} = M_{0i}(t) - M_i(t). \quad (2.1)$$

Here $M_i(t)$ is the i th particle magnetization; M_{0i} is the equilibrium magnetization in the field, acting on the particle at the given moment t ; τ is the Néel relaxation time, which can be estimated as $\tau \sim \tau_0 \exp(KV/k_B T)$ [20]; k_B is the Boltzmann constant; T is the absolute temperature; V is the

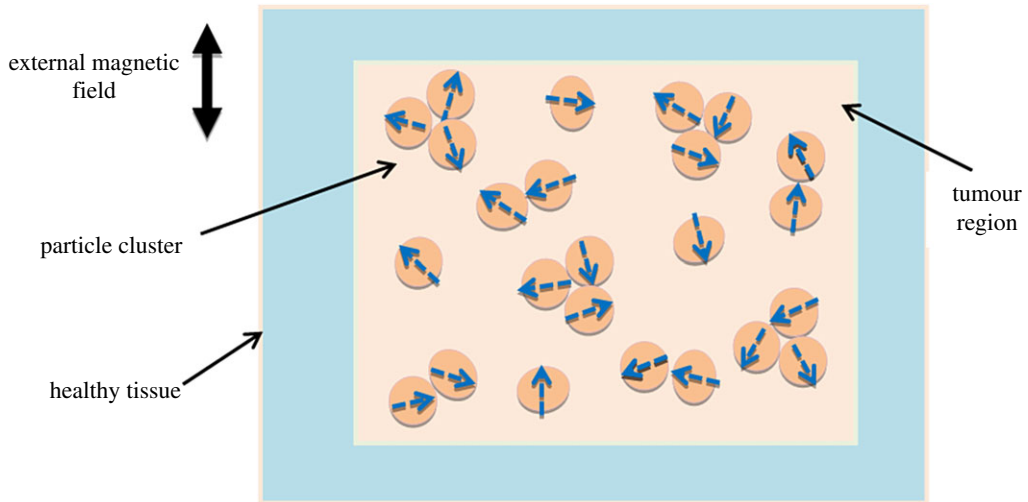


Figure 1. Sketch of the considered clusters of ferromagnetic particles. Dashed arrows illustrate the particles' magnetic moments. (Online version in colour.)

particle volume; and τ_0 is the time of the Larmor precession of the particle magnetic moment. For iron oxide particles, $\tau_0 \sim 10^{-9}$ s; $K \approx 14$ kJ m $^{-3}$ (e.g. [21]).

Simple estimates show that for magnetite particles with diameter 20 nm the anisotropy energy KV is about 15 times more than the thermal energy $k_B T$; therefore, the strong inequality $KV \gg k_B T$ holds. For particles with diameter less than 8 nm, the opposite inequality $KV < k_B T$ is true.

For maximal simplification of calculations, in order to concentrate attention on the effect of particle clusterization, we will consider the limiting case of relatively weak fields, when the Zeeman energy of the particle interaction with the heating field H is less than the thermal energy $k_B T$. The linear relation between the particle magnetization and the field can be used in this situation. Next, for maximal simplification of calculations, to get results in a physically transparent form, we will consider the two-dimensional approximation, assuming that the particle centres, their axes of easy magnetization and the applied magnetic field are in the same plane.

(b) Intensity of heat production

The intensity of energy adsorption by each of the particles can be determined from the general relation of thermodynamics of magnetizable media (e.g. [15]):

$$W = -\mu_0 \frac{V}{\Theta} \int_0^\Theta \left(M(t) \cdot \frac{dH_e}{dt} \right) dt. \quad (2.2)$$

Here $H_e = H_0 \cos \omega t$ is the external alternating magnetic field; $\Theta = 2\pi/\omega$ is the field period.

Equation (2.2) can be transformed as

$$W = \mu_0 \frac{\omega^2 V}{2\pi} \int_0^{2\pi/\omega} (M \cdot H_0) \sin \omega t \, dt. \quad (2.3)$$

The averaged, over the N -particle cluster, particle magnetization M is

$$M = \frac{1}{N} \sum_{i=1}^N M_i e^{i\omega t}, \quad (2.4)$$

where the i th particle magnetization M_i can be determined as a solution of the system (2.1).

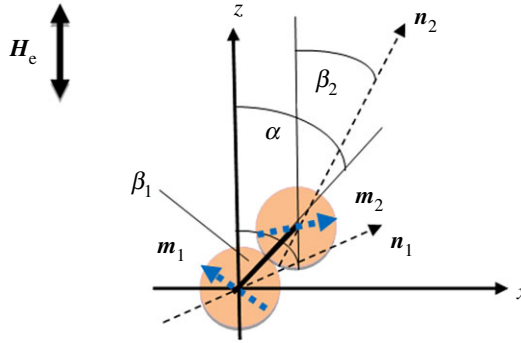


Figure 2. Sketch of doublet particles. $\mathbf{H}_e = \mathbf{H}_0 \cos \omega t$. (Online version in colour.)

3. Particle clusters with strong magnetic anisotropy

The magnetization \mathbf{M}_i of the magnetically strongly anisotropic particle must with high probability be oriented either parallel or antiparallel to the axis of easy magnetization of the particle. In the linear approximation with respect to the acting field, the particle equilibrium magnetization can be represented as

$$\mathbf{M}_{0i} = \chi_0 n_i (\mathbf{H}_i^{\text{eff}} \cdot \mathbf{n}_i). \quad (3.1)$$

Here, $\mathbf{H}_i^{\text{eff}}$ is the magnetic field acting on the i th particle; $\chi_0 = \mu_0 M_s^2 V / k_B T$; μ_0 is the vacuum permeability; M_s is the saturation magnetization of the particle material; \mathbf{n}_i is the unit vector of the axis of easy magnetization of the particle; and the subscript i marks the particle in the cluster. We can express $\mathbf{H}_i^{\text{eff}}$ and \mathbf{n}_i in the coordinate system (x, z) as $\mathbf{H}_i^{\text{eff}} = (H_{x_i}^{\text{eff}}, H_{z_i}^{\text{eff}})$ and $\mathbf{n}_i = (n_{x_i}, n_{z_i})$, respectively.

Combining equations (2.1) and (3.1), we come to the following equations for the components of the particle magnetization:

$$\left. \begin{aligned} \tau \frac{dM_{x_i}(t)}{dt} + M_{x_i}(t) &= \chi_0 n_{x_i} (H_{x_i}^{\text{eff}} n_{x_i} + H_{z_i}^{\text{eff}} n_{z_i}) \\ \tau \frac{dM_{z_i}(t)}{dt} + M_{z_i}(t) &= \chi_0 n_{z_i} (H_{x_i}^{\text{eff}} n_{x_i} + H_{z_i}^{\text{eff}} n_{z_i}) \end{aligned} \right\} \quad (3.2)$$

and

Here the right sides of equations (3.2) are the components of equilibrium magnetization of the particle in the field $\mathbf{H}_i^{\text{eff}}$; the components $H_{x_i}^{\text{eff}}$ and $H_{z_i}^{\text{eff}}$ will be determined in the next sections.

(a) Model of a pair of particles (a doublet)

In this subsection, we will consider the two-particle cluster (doublet), illustrated in figure 2. Let us denote the angle between the doublet axis and the applied field as α ; and the angle between the vector \mathbf{n}_i of the i th particle axis of easy magnetization and the direction of the external magnetic field \mathbf{H}_e as β_i .

The components of unit vectors \mathbf{n}_i of easy magnetization can be represented in the form (figure 2)

$$n_{x_i} = \sin \beta_i, \quad n_{z_i} = \cos \beta_i \quad (i = 1, 2). \quad (3.3)$$

Combining relations (3.2) and (3.3), we come to the following system of equations for the components of the particles' magnetization:

$$\left. \begin{aligned} \tau \frac{dM_{x_1}}{dt} + M_{x_1} &= \chi_0(H_{x_1}^{\text{eff}} \sin^2 \beta_1 + H_{z_1}^{\text{eff}} \sin \beta_1 \cos \beta_1), \\ \tau \frac{dM_{z_1}}{dt} + M_{z_1} &= \chi_0(H_{x_1}^{\text{eff}} \sin \beta_1 \cos \beta_1 + H_{z_1}^{\text{eff}} \cos^2 \beta_1), \\ \tau \frac{dM_{x_2}}{dt} + M_{x_2} &= \chi_0(H_{x_2}^{\text{eff}} \sin^2 \beta_2 + H_{z_2}^{\text{eff}} \sin \beta_2 \cos \beta_2), \\ \tau \frac{dM_{z_2}}{dt} + M_{z_2} &= \chi_0(H_{x_2}^{\text{eff}} \sin \beta_2 \cos \beta_2 + H_{z_2}^{\text{eff}} \cos^2 \beta_2). \end{aligned} \right\} \quad (3.4)$$

and

The field H_i^{eff} can be written down in the form

$$\left. \begin{aligned} H_i^{\text{eff}} &= H_0 \cos \omega t + H_i^{\text{d}} \\ H_i^{\text{d}} &= \frac{1}{4\pi} \left(\frac{3(\mathbf{m}_j \cdot \mathbf{r})\mathbf{r} - r^2 \mathbf{m}_j}{r^5} \right), \end{aligned} \right\} \quad (3.5)$$

with

where H_i^{d} is the field of the dipole–dipole interaction between the i th and j th particles in the aggregate ($i, j = 1, 2$ and $i \neq j$); \mathbf{r} is the radius vector, connecting the centres of the particles; and $\mathbf{m}_j = V\mathbf{M}_j$ is the particle magnetic moment.

The second relation in equation (3.5) can be rewritten as

$$H_{ij}^{\text{d}} = \frac{V}{4\pi} \left(\frac{3(\mathbf{M}_j \cdot \mathbf{r})\mathbf{r} - r^2 \mathbf{M}_j}{r^5} \right), \quad (3.6)$$

with

$$\mathbf{M}_j = (M_{x_j}, M_{z_j})$$

and

$$\mathbf{r} = (r_x, r_z) = d(\sin \alpha, \cos \alpha),$$

where d is the particle diameter. Combining (3.5) and (3.6), we come to the following relation for the components of the effective magnetic field H_i^{eff} :

$$\left. \begin{aligned} H_{x_i}^{\text{eff}} &= \frac{1}{24}((3 \sin^2 \alpha - 1)M_{x_j} + 3 \cos \alpha \sin \alpha M_{z_j}), \\ H_{z_i}^{\text{eff}} &= H_0 \cos \omega t + \frac{1}{24}((3 \cos^2 \alpha - 1)M_{z_j} + 3 \cos \alpha \sin \alpha M_{x_j}), \end{aligned} \right\} \quad (3.7)$$

with

$$i, j = 1, 2, \quad i \neq j.$$

Substituting (3.7) into equation (3.4), we get the following system of equations for the magnetization dynamics:

$$\begin{aligned} \tau \frac{dM_{x_1}}{dt} + M_{x_1} - \frac{\chi_0}{24} [&((3 \sin^2 \alpha - 1)M_{x_2} + 3 \cos \alpha \sin \alpha M_{z_2}) \sin^2 \beta_1 \\ &+ (3 \cos \alpha \sin \alpha M_{x_2} + (3 \cos^2 \alpha - 1)M_{z_2}) \sin \beta_1 \cos \beta_1] \\ &= \chi_0 H_0 \sin \beta_1 \cos \beta_1 \cos \omega t, \end{aligned}$$

$$\begin{aligned}
& \tau \frac{dM_{x_2}}{dt} + M_{x_2} - \frac{\chi_0}{24} [((3 \sin^2 \alpha - 1)M_{x_1} + 3 \cos \alpha \sin \alpha M_{z_1}) \sin^2 \beta_2 \\
& \quad + (3 \cos \alpha \sin \alpha M_{x_1} + (3 \cos^2 \alpha - 1)M_{z_1}) \sin \beta_2 \cos \beta_2] \\
& = \chi_0 H_0 \sin \beta_2 \cos \beta_2 \cos \omega t,
\end{aligned} \tag{3.8}$$

$$\begin{aligned}
& \tau \frac{dM_{z_1}}{dt} + M_{z_1} - \frac{\chi_0}{24} [((3 \cos^2 \alpha - 1)M_{z_2} + 3 \cos \alpha \sin \alpha M_{x_2}) \cos^2 \beta_1 \\
& \quad + (3 \cos \alpha \sin \alpha M_{z_2} + (3 \sin^2 \alpha - 1)M_{x_2}) \sin \beta_1 \cos \beta_1] \\
& = \chi_0 H_0 \cos^2 \beta_1 \cos \omega t
\end{aligned}$$

and

$$\begin{aligned}
& \tau \frac{dM_{z_2}}{dt} + M_{z_2} - \frac{\chi_0}{24} [((3 \cos^2 \alpha - 1)M_{z_1} + 3 \cos \alpha \sin \alpha M_{x_1}) \cos^2 \beta_2 \\
& \quad + (3 \cos \alpha \sin \alpha M_{z_1} + (3 \sin^2 \alpha - 1)M_{x_1}) \sin \beta_1 \cos \beta_1] \\
& = \chi_0 H_0 \cos^2 \beta_1 \cos \omega t.
\end{aligned}$$

Relations (3.8) represent a system of linear differential equations with respect to the components of the vectors M_j ($j = 1, 2$). To solve it, we use the standard mathematical approach, changing $\cos \omega t$ to $\exp(i\omega t)$. In the frame of this approach, one can present $M_j = M_j^0 \exp(i\omega t)$, where M_j^0 are complex magnetization amplitudes, to be determined. By using these forms in equation (3.8), we come to the following system of algebraic equations for the complex amplitudes:

$$\begin{aligned}
& (1 + i\omega\tau)M_{x_1}^0 - \frac{\chi_0}{24} [((3 \sin^2 \alpha - 1)M_{x_2}^0 + 3 \cos \alpha \sin \alpha M_{z_2}^0) \sin^2 \beta_1 \\
& \quad + (3 \cos \alpha \sin \alpha M_{x_2}^0 + (3 \cos^2 \alpha - 1)M_{z_2}^0) \sin \beta_1 \cos \beta_1] \\
& = \chi_0 H_0 \sin \beta_1 \cos \beta_1, \\
& (1 + i\omega\tau)M_{x_2}^0 - \frac{\chi_0}{24} [((3 \sin^2 \alpha - 1)M_{x_1}^0 + 3 \cos \alpha \sin \alpha M_{z_1}^0) \sin^2 \beta_2 \\
& \quad + (3 \cos \alpha \sin \alpha M_{x_1}^0 + (3 \cos^2 \alpha - 1)M_{z_1}^0) \sin \beta_2 \cos \beta_2] \\
& = \chi_0 H_0 \sin \beta_2 \cos \beta_2,
\end{aligned} \tag{3.9}$$

$$\begin{aligned}
& (1 + i\omega\tau)M_{z_1}^0 - \frac{\chi_0}{24} [((3 \cos^2 \alpha - 1)M_{z_2}^0 + 3 \cos \alpha \sin \alpha M_{x_2}^0) \cos^2 \beta_1 \\
& \quad + (3 \cos \alpha \sin \alpha M_{z_2}^0 + (3 \sin^2 \alpha - 1)M_{x_2}^0) \sin \beta_1 \cos \beta_1] \\
& = \chi_0 H_0 \cos^2 \beta_1
\end{aligned}$$

and

$$\begin{aligned}
& (1 + i\omega\tau)M_{z_2}^0 - \frac{\chi_0}{24} [((3 \cos^2 \alpha - 1)M_{z_1}^0 + 3 \cos \alpha \sin \alpha M_{x_1}^0) \cos^2 \beta_2 \\
& \quad + (3 \cos \alpha \sin \alpha M_{z_1}^0 + (3 \sin^2 \alpha - 1)M_{x_1}^0) \sin \beta_1 \cos \beta_1] \\
& = \chi_0 H_0 \cos^2 \beta_1.
\end{aligned}$$

The solution of the system (3.9) gives us the amplitudes $M_{z_j}^0 = (M_{z_j}^0)' + i(M_{z_j}^0)''$ and $M_{x_j}^0 = (M_{x_j}^0)' + i(M_{x_j}^0)''$.

Since the angles $\beta_{1,2}$ and α have arbitrary values, the physically determined (measured) value of the intensity of heat production can be calculated as

$$\langle W \rangle = \frac{1}{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} W(\beta_1, \beta_2) f(\beta_1) f(\beta_2) d\beta_1 d\beta_2 d\alpha. \tag{3.10}$$

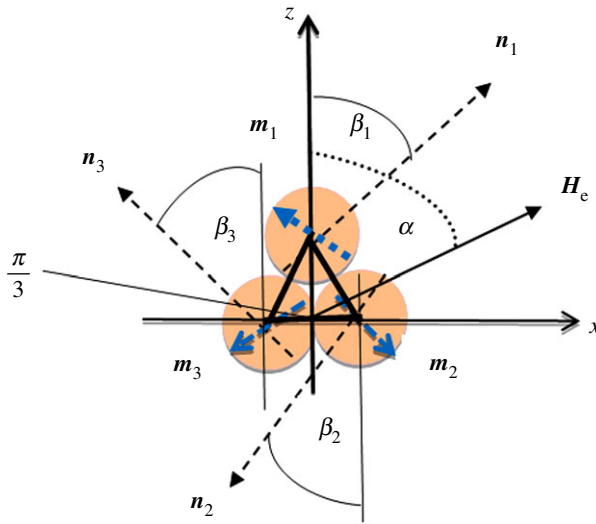


Figure 3. Sketch of the cluster of three nanoparticles; H_e is the applied magnetic field; H_e is inclined with respect to the axis z at angle α . (Online version in colour.)

Here $f(\beta_j)$ is the probability density of the given orientation of the j th particle axis of easy magnetization. If the axes have the most energetically favourable orientations,

$$f(\beta_j) = \delta(\beta_j - \alpha). \quad (3.11)$$

In the case of random orientation of the axes,

$$f(\beta_j) = \frac{1}{2\pi}. \quad (3.12)$$

Only the term with the imaginary part of the solution of $M_{z_j}^0$ will give a non-zero contribution to the intensity of heat production. The result reads

$$\langle W \rangle = -\frac{1}{2\pi} \mu_0 V H_0 \omega \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \left(\frac{\sum_{j=1}^2 (M_{z_j}^0(\beta_1, \beta_2, \alpha))''}{2} \right) f(\beta_1) f(\beta_2) d\beta_1 d\beta_2 d\alpha. \quad (3.13)$$

If the particles, axes directions correspond to the most favourable state (i.e. the relation (3.11) holds), one gets

$$\langle W \rangle = -\frac{1}{2\pi} \mu_0 V H_0 \omega \int_0^{2\pi} \left(\frac{\sum_{j=1}^2 (M_{z_j}^0(\alpha))''}{2} \right) d\alpha. \quad (3.14)$$

In the case of random orientation of the axes, corresponding to equation (3.12), one has

$$\langle W \rangle = -\frac{1}{16\pi^3} \mu_0 V H_0 \omega \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \left(\frac{\sum_{j=1}^2 (M_{z_j}^0(\beta_1, \beta_2, \alpha))''}{2} \right) d\beta_1 d\beta_2 d\alpha. \quad (3.15)$$

(b) Model of three-particle cluster

Let us consider the three-particle cluster, presented in figure 3. The particles' axes of easy magnetization have an arbitrary orientation.

The components of unit vectors \mathbf{n}_i of easy magnetization can be represented as

$$\begin{aligned} n_{x_1} &= \sin \beta_1, & n_{z_1} &= \cos \beta_1, \\ n_{x_2} &= -\sin \beta_2, & n_{z_1} &= -\cos \beta_2, \\ n_{x_1} &= -\sin \beta_3, & n_{z_1} &= \cos \beta_3. \end{aligned} \quad (3.16)$$

Combining relations (2.3) and (3.16), the system of equations of remagnetization becomes

$$\left. \begin{aligned} \tau \frac{dM_{x_1}}{dt} + M_{x_1} &= \chi_0 (H_{x_1}^{\text{eff}} \sin^2 \beta_1 + H_{z_1}^{\text{eff}} \sin \beta_1 \cos \beta_1), \\ \tau \frac{dM_{z_1}}{dt} + M_{z_1} &= \chi_0 (H_{x_1}^{\text{eff}} \sin \beta_1 \cos \beta_1 + H_{z_1}^{\text{eff}} \cos^2 \beta_1), \\ \tau \frac{dM_{x_2}}{dt} + M_{x_2} &= \chi_0 (H_{x_2}^{\text{eff}} \sin^2 \beta_2 + H_{z_2}^{\text{eff}} \sin \beta_2 \cos \beta_2), \\ \tau \frac{dM_{z_2}}{dt} + M_{z_2} &= \chi_0 (H_{x_2}^{\text{eff}} \sin \beta_2 \cos \beta_2 + H_{z_2}^{\text{eff}} \cos^2 \beta_2), \\ \tau \frac{dM_{x_3}}{dt} + M_{x_3} &= \chi_0 (H_{x_3}^{\text{eff}} \sin^2 \beta_3 - H_{z_3}^{\text{eff}} \sin \beta_3 \cos \beta_3) \\ \text{and} \quad \tau \frac{dM_{z_3}}{dt} + M_{z_3} &= \chi_0 (-H_{x_3}^{\text{eff}} \sin \beta_3 \cos \beta_3 + H_{z_3}^{\text{eff}} \cos^2 \beta_3). \end{aligned} \right\} \quad (3.17)$$

The effective field $\mathbf{H}_i^{\text{eff}}$, acting on the i th particle in the cluster, again can be expressed in the form

$$\begin{aligned} \mathbf{H}_i^{\text{eff}} &= \mathbf{H}_e + \mathbf{H}_i^{\text{d}}, \\ \mathbf{H}_i^{\text{d}} &= \mathbf{H}_{ij}^{\text{d}} + \mathbf{H}_{il}^{\text{d}}. \end{aligned} \quad (3.18)$$

Here again \mathbf{H}_e is the oscillating field, external with respect to the cluster; \mathbf{H}_i^{d} is the field of the dipole–dipole interaction of the i th particle of the cluster with the j th and l th ones ($i, j, l = 1, 2, 3$ and $i \neq j \neq l$); and \mathbf{r}_{ij} , \mathbf{r}_{il} are the radius vectors, connecting the centres of the corresponding particles. The two terms on the right side of the second relation in (3.18) can be represented as

$$\begin{aligned} \mathbf{H}_{ij}^{\text{d}} &= \frac{V}{4\pi} \left(\frac{3(\mathbf{M}_j \cdot \mathbf{r}_{ij})\mathbf{r}_{ij} - M_j r_{ij}^2}{r_{ij}^5} \right), & \mathbf{H}_{il}^{\text{d}} &= \frac{V}{4\pi} \left(\frac{3(\mathbf{M}_l \cdot \mathbf{r}_{il})\mathbf{r}_{il} - M_l r_{il}^2}{r_{il}^5} \right), \\ \mathbf{M}_j &= (M_{x_j}, M_{z_j}), & \mathbf{M}_l &= (M_{x_l}, M_{z_l}), \end{aligned} \quad (3.19)$$

and

$$\begin{aligned} \mathbf{r}_{12} &= (r_{x12}, r_{z12}) = \left(d \cos \frac{\pi}{3}, -d \sin \frac{\pi}{3} \right) = -\mathbf{r}_{21}, \\ \mathbf{r}_{23} &= (r_{x23}, r_{z23}) = (-d, 0) = -\mathbf{r}_{32}, \\ \mathbf{r}_{31} &= (r_{x31}, r_{z31}) = \left(d \cos \frac{\pi}{3}, d \sin \frac{\pi}{3} \right) = -\mathbf{r}_{13}. \end{aligned}$$

The three-particle cluster, shown in figure 3, can have an arbitrary orientation with respect to the external field \mathbf{H}_e . The physical meaning has the intensity of heat production averaged over these orientations. For calculations, it is more convenient to fix the 'triangle' orientation and to average over the relative orientations of the field \mathbf{H}_e . In figure 3, these field components are

$$H_{ex} = H_0 \sin \alpha \cos \omega t \quad \text{and} \quad H_{ez} = H_0 \cos \alpha \cos \omega t. \quad (3.20)$$

Substituting relations (3.18) and (3.20) into (3.17), one gets

$$\left. \begin{aligned} & \tau \frac{dM_{x_1}}{dt} + M_{x_1} - \chi_0(H_{x_1}^d \sin^2 \beta_1 + H_{z_1}^d \sin \beta_1 \cos \beta_1) \\ & \quad = \chi_0 H_0 \cos \omega t (\sin \alpha \sin^2 \beta_1 + \cos \alpha \sin \beta_1 \cos \beta_1), \\ & \tau \frac{dM_{z_1}}{dt} + M_{z_1} - \chi_0(H_{x_1}^d \sin \beta_1 \cos \beta_1 + H_{z_1}^d \cos^2 \beta_1) \\ & \quad = \chi_0 H_0 \cos \omega t (\sin \alpha \sin \beta_1 \cos \beta_1 + \cos \alpha \cos^2 \beta_1), \\ & \tau \frac{dM_{x_2}}{dt} + M_{x_2} - \chi_0(H_{x_2}^d \sin^2 \beta_2 + H_{z_2}^d \sin \beta_2 \cos \beta_2) \\ & \quad = \chi_0 H_0 \cos \omega t (\sin \alpha \sin^2 \beta_2 + \cos \alpha \sin \beta_2 \cos \beta_2), \\ & \tau \frac{dM_{z_2}}{dt} + M_{z_2} - \chi_0(H_{x_2}^d \sin \beta_2 \cos \beta_2 + H_{z_2}^d \cos^2 \beta_2) \\ & \quad = \chi_0 H_0 \cos \omega t (\sin \alpha \sin \beta_2 \cos \beta_2 + \cos \alpha \cos^2 \beta_2), \\ & \tau \frac{dM_{x_3}}{dt} + M_{x_3} - \chi_0(H_{x_3}^d \sin^2 \beta_3 - H_{z_3}^d \sin \beta_3 \cos \beta_3) \\ & \quad = \chi_0 H_0 \cos \omega t (\sin \alpha \sin^2 \beta_3 - \cos \alpha \sin \beta_3 \cos \beta_3) \\ \text{and} \quad & \tau \frac{dM_{z_3}}{dt} + M_{z_3} - \chi_0(-H_{x_3}^d \sin \beta_3 \cos \beta_3 + H_{z_3}^d \cos^2 \beta_3) \\ & \quad = \chi_0 H_0 \cos \omega t (-\sin \alpha \sin \beta_3 \cos \beta_3 + \cos \alpha \cos^2 \beta_3). \end{aligned} \right\} \quad (3.21)$$

By using (3.19), we can represent the second relation in (3.18) as

$$\left. \begin{aligned} H_{x_1}^d &= \frac{1}{96} (-M_{x_2} - M_{x_3} - 3\sqrt{3}M_{z_2} + 3\sqrt{3}M_{z_3}), \\ H_{z_1}^d &= \frac{1}{96} (-3\sqrt{3}M_{x_2} + 3\sqrt{3}M_{x_3} + 5M_{z_2} + 5M_{z_3}), \\ H_{x_2}^d &= \frac{1}{96} (8M_{x_3} - M_{x_1} - 3\sqrt{3}M_{z_1}), \\ H_{z_2}^d &= \frac{1}{96} (-3\sqrt{3}M_{x_1} - 4M_{z_3} + 5M_{z_1}), \\ H_{x_3}^d &= \frac{1}{96} (-M_{x_1} + 8M_{x_2} + 3\sqrt{3}M_{z_1}) \\ \text{and} \quad & H_{z_3}^d = \frac{1}{96} (3\sqrt{3}M_{x_1} + 5M_{z_1} - 4M_{z_2}). \end{aligned} \right\} \quad (3.22)$$

Substituting equations (3.22) into equations (3.21), we come to the system of equations for components of magnetization of the particles in the cluster:

$$\left. \begin{aligned}
 & \tau \frac{dM_{x_1}}{dt} + M_{x_1} - \frac{\chi_0}{96} \left((-M_{x_2} - M_{x_3} - 3\sqrt{3}M_{z_2} + 3\sqrt{3}M_{z_3}) \sin^2 \beta_1 \right. \\
 & \quad \left. + (-3\sqrt{3}M_{x_2} + 3\sqrt{3}M_{x_3} + 5M_{z_2} + 5M_{z_3}) \sin \beta_1 \cos \beta_1 \right) \\
 & = \chi_0 H_0 \cos \omega t (\sin \alpha \sin^2 \beta_1 + \cos \alpha \sin \beta_1 \cos \beta_1), \\
 & \tau \frac{dM_{z_1}}{dt} + M_{z_1} - \frac{\chi_0}{96} \left((-M_{x_2} - M_{x_3} - 3\sqrt{3}M_{z_2} + 3\sqrt{3}M_{z_3}) \sin \beta_1 \cos \beta_1 \right. \\
 & \quad \left. + (-3\sqrt{3}M_{x_2} + 3\sqrt{3}M_{x_3} + 5M_{z_2} + 5M_{z_3}) \cos^2 \beta_1 \right) \\
 & = \chi_0 H_0 \cos \omega t (\sin \alpha \sin \beta_1 \cos \beta_1 + \cos \alpha \cos^2 \beta_1), \\
 & \tau \frac{dM_{x_2}}{dt} + M_{x_2} - \frac{\chi_0}{96} \left((8M_{x_3} - M_{x_1} - 3\sqrt{3}M_{z_1}) \sin^2 \beta_2 \right. \\
 & \quad \left. + (-3\sqrt{3}M_{x_1} - 4M_{z_3} + 5M_{z_1}) \sin \beta_2 \cos \beta_2 \right) \\
 & = \chi_0 H_0 \cos \omega t (\sin \alpha \sin^2 \beta_2 + \cos \alpha \sin \beta_2 \cos \beta_2), \\
 & \tau \frac{dM_{z_2}}{dt} + M_{z_2} - \frac{\chi_0}{96} \left((8M_{x_3} - M_{x_1} - 3\sqrt{3}M_{z_1}) \sin \beta_2 \cos \beta_2 \right. \\
 & \quad \left. + (-3\sqrt{3}M_{x_1} - 4M_{z_3} + 5M_{z_1}) \cos^2 \beta_2 \right) \\
 & = \chi_0 H_0 \cos \omega t (\sin \alpha \sin \beta_2 \cos \beta_2 + \cos \alpha \cos^2 \beta_2), \\
 & \tau \frac{dM_{x_3}}{dt} + M_{x_3} - \frac{\chi_0}{96} \left((-M_{x_1} + 8M_{x_2} + 3\sqrt{3}M_{z_1}) \sin^2 \beta_3 \right. \\
 & \quad \left. - (3\sqrt{3}M_{x_1} + 5M_{z_1} - 4M_{z_2}) \sin \beta_3 \cos \beta_3 \right) \\
 & = \chi_0 H_0 \cos \omega t (\sin \alpha \sin^2 \beta_3 - \cos \alpha \sin \beta_3 \cos \beta_3) \\
 \text{and} \quad & \tau \frac{dM_{z_3}}{dt} + M_{z_3} - \frac{\chi_0}{96} \left((-M_{x_1} + 8M_{x_2} + 3\sqrt{3}M_{z_1}) \sin \beta_3 \cos \beta_3 \right. \\
 & \quad \left. + (3\sqrt{3}M_{x_1} + 5M_{z_1} - 4M_{z_2}) \cos^2 \beta_3 \right) \\
 & = \chi_0 H_0 \cos \omega t (-\sin \alpha \sin \beta_3 \cos \beta_3 + \cos \alpha \cos^2 \beta_3).
 \end{aligned} \right\} \quad (3.23)$$

Relations (3.23) represent a system of Debye equations with respect to the components of the vectors M_j . To solve it, again we change $\cos \omega t$ to $\exp(i\omega t)$. In the frame of this approach, one can represent $M_j = M_j^0 \exp(i\omega t)$, where M_j^0 are magnetization amplitudes, to be determined. By using these forms in equations (3.23), we will come to an algebraic system of equations. The solution of the algebraic system of equations gives us the complex amplitudes $M_{z_i}^0 = (M_{z_i}^0)' + i(M_{z_i}^0)''$ and $M_{x_i}^0 = (M_{x_i}^0)' + i(M_{x_i}^0)''$. The obtained solution of the equations will be used to estimate the intensity of heat production at an arbitrary orientation of the easy magnetization axes.

In the general case, we can represent the MH heat production averaged over all orientation angles β_i of easy magnetization axes of the nanoparticles in the cluster as

$$\langle W \rangle = \frac{1}{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} W(\beta_1, \beta_2, \beta_3, \alpha) f(\beta_1) f(\beta_2) f(\beta_3) d\beta_1 d\beta_2 d\beta_3 d\alpha, \quad (3.24)$$

where $f(\beta_i)$ again is the distribution function for the angle β_i .

On the basis of (2.2), (2.4) and (3.24), only terms with the imaginary part of the solution of the system of equations (3.23) will give a non-zero contribution to the intensity of heat production.

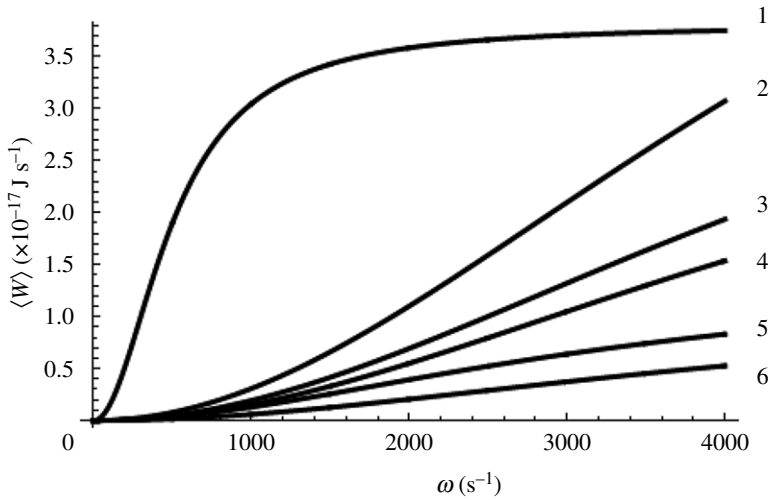


Figure 4. Intensity of heat production $\langle W \rangle$ per particle versus the field frequency ω . Figures near the curves: 1, single particle with chaotic orientation of its axis of easy magnetization; 2, chain of two particles aligned along magnetic field (in equation (3.14) $\alpha = \beta_1 = \beta_2 \equiv 0$); 3, doublet particles with random orientation of the particle axes and the doublet orientation (α, β_1, β_2 have arbitrary, equal probable values in equation (3.15)); 4, doublet particles with the most energetically profitable orientation ‘head to tail’ ($\beta_1 = \beta_2 = \alpha$ in equation (3.14)) and random distribution for α ; 5, cluster of three particles with random orientation of easy magnetization axes and the field \mathbf{H}_e with respect to the cluster (in equation (3.25) $\alpha, \beta_1, \beta_2, \beta_3$ have arbitrary values); 6, cluster of three particles with the most energetically profitable configuration of the vectors of easy magnetization axes (in equation (3.25) $\beta_1 \equiv \pi/2, \beta_2 \equiv \pi/3, \beta_3 \equiv \pi/3$ and random distribution for the orientation angle α of the cluster with respect to the field). The diameter of particles is 20 nm. Values of the system’s physical parameters: $\tau \approx 2 \times 10^{-3} \text{ s}$; $M_s = 4.5 \times 10^5 \text{ A m}^{-1}$; $H_0 = 15 \times 10^3 \text{ A m}^{-1}$; $T = 300^\circ \text{ K}$.

The result reads

$$\left. \begin{aligned} \langle W \rangle = \frac{1}{2\pi} \mu_0 V H_0 \omega \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \int_0^{2\pi} \left(-\frac{1}{N} \left(\sum_{i=1}^3 \left(\frac{\mathbf{M}_i(\beta_1, \beta_2, \beta_3, \alpha) \cdot \mathbf{H}_e}{|\mathbf{H}_e|} \right) \right) \right) \\ \times f(\beta_1) f(\beta_2) f(\beta_3) d\beta_1 d\beta_2 d\beta_3 d\alpha, \\ N = 3. \end{aligned} \right\} \quad (3.25)$$

Here $|\mathbf{H}_e|$ is the absolute value of magnetic field \mathbf{H}_e that is expressed in (3.20).

Some results of MH heat production $\langle W \rangle$ are illustrated in figure 4 for a single particle as well as for clusters of two and three particles.

The obtained results demonstrate that clusterization of the particles with strong magnetic anisotropy decreases the thermal effect during MH. The MH heat production for the random orientation of the particles’ axes of easy magnetization is more than that in the cases of the most energetically favourable orientations. These conclusions are in agreement with the experimental results of [9] where irregular-shaped aggregates consisting of many particles were observed.

4. Particle clusters with weak anisotropy

In this section, we consider the limiting case of zero magnetic anisotropy of the particles. Physically, this means that the size of the particles is small and the energy of their magnetic

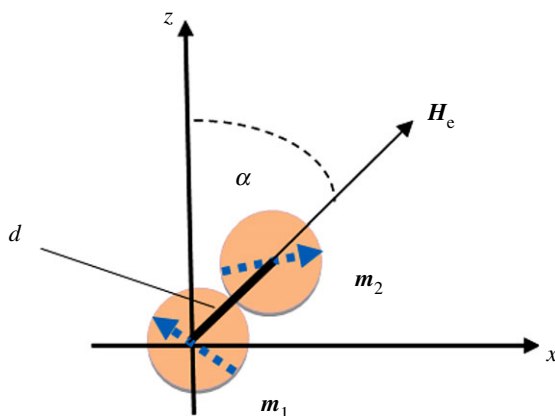


Figure 5. Sketch of the two-particle cluster. H_e is inclined with respect to the axis z at angle α .

anisotropy KV is less than the thermal energy $k_B T$. Let us denote the field acting on the particle H_e . In this field, the particle equilibrium magnetization is

$$M_{0i} = \frac{H_e}{H} \frac{m}{V} L\left(\mu_0 \frac{mH}{k_B T}\right), \quad H = |H_e|. \quad (4.1)$$

Here m is the absolute value of the particle magnetic moment, V is its volume and $L(\mu_0(mH_e/k_B T))$ is the Langevin function. For simplicity again, we consider the case of relatively weak fields and suppose that $\mu_0(mH_e/k_B T) \ll 1$. In this situation, instead of (4.1) one gets

$$M_{0i} = \frac{1}{3} \frac{H_e}{H} \frac{m}{V} \mu_0 \frac{mH_e}{k_B T}. \quad (4.2)$$

If the particle is in a cluster, equation (4.2) transforms to

$$\left. \begin{aligned} M_{0i} &= \kappa H_i^{\text{eff}}, \quad \kappa = \frac{1}{3} \mu_0 \frac{m^2}{k_B T V}, \\ H_i^{\text{eff}} &= H_e + H_i^{\text{d}} \\ H_i^{\text{d}} &= \frac{V}{4\pi} \sum_{j \neq i}^N \frac{3r_{ij}(r_{ij} \cdot M_j) - r_{ij}^2 M_j}{r_{ij}^5} \end{aligned} \right\} \quad (4.3)$$

and

Here again H_e is an external (applied) magnetic field, i, j are numbers of the particles in the cluster, r_{ij} is the radius vector between the centres of the corresponding particles in the cluster and N is the number of particles inside the cluster. The summation is over all interaction particles j in the cluster where $j = 1, 2, \dots, N$. It will be convenient to use the coordinate system (x, z) with the axis Oz aligned along the doublet axis, illustrated in figures 5 and 6. In this coordinate system, $H_e = H_0 \cos \omega t (\sin \alpha, \cos \alpha)$ and angle α will be discussed in the next sections (figures 5 and 6).

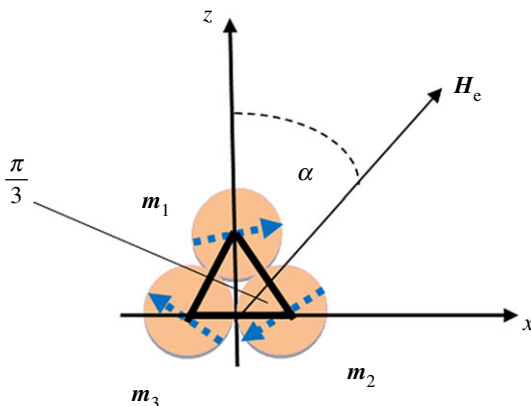


Figure 6. Sketch of the three-nanoparticle cluster. (Online version in colour.)

(a) Model of chain-like two particles

We suppose that the particle magnetic anisotropy is negligible. The relations (4.3) can be represented as

$$\begin{aligned} M_{0i} &= \kappa(H_e + H_i^d), \\ H_i^d &= \frac{V}{4\pi} \left(\frac{3\mathbf{r} \cdot \mathbf{M}_j - r^2 M_j}{r^5} \right) \end{aligned} \quad (4.4)$$

with

$$i, j = 1, 2.$$

Here \mathbf{r} is the radius vector between the centres of the particles in the chain. In the coordinate system, presented in figure 5, $\mathbf{M}_j = (M_{xj}, M_{zj})$.

Combining equations (2.1), (3.20) and (4.4), we come to the following system of equations for the components of the particles' magnetization:

$$\left. \begin{aligned} \tau \frac{dM_{x_i}}{dt} + M_{x_i} + \frac{\kappa}{24} M_{x_j} &= \kappa H_0 \cos \omega t \sin \alpha, \\ \tau \frac{dM_{z_i}}{dt} + M_{z_i} - \frac{\kappa}{12} M_{z_j} &= \kappa H_0 \cos \omega t \cos \alpha \end{aligned} \right\} \quad (4.5)$$

with

$$i, j = 1, 2 \text{ and } i \neq j.$$

For the simplicity of calculations, we again use in (4.5) the complex exponent $\exp(i\omega t)$ instead of $\cos \omega t$ and obtain the components of magnetization \mathbf{M}_i in the complex form $\mathbf{M}_i = [(M_i^0)' + i(M_i^0)'] \exp(i\omega t)$. Finally, one gets the heat production for a two-particle cluster over average orientation of chain-like two particles around angle α

$$\langle W \rangle = \frac{1}{4\pi} \mu_0 V H_0 \omega \int_0^{2\pi} \left(-\frac{1}{2} \sum_{i=1}^2 ((M_{x_i}^0(\alpha))'' \sin \alpha + (M_{z_i}^0(\alpha))'' \cos \alpha) \right) d\alpha. \quad (4.6)$$

(b) Three-particle cluster

In this section, we consider the three-particle cluster, illustrated in figure 6. The acting field $\mathbf{H}_i^{\text{eff}}$ can be expressed as

$$\mathbf{H}_i^{\text{eff}} = \mathbf{H}_e + \frac{V}{4\pi} \left(\frac{3(\mathbf{M}_j \cdot \mathbf{r}_{ij})\mathbf{r}_{ij} - M_j r_{ij}^2}{r_{ij}^5} \right) + \frac{V}{4\pi} \left(\frac{3(\mathbf{M}_l \cdot \mathbf{r}_{il})\mathbf{r}_{il} - M_l r_{il}^2}{r_{il}^5} \right). \quad (4.7)$$

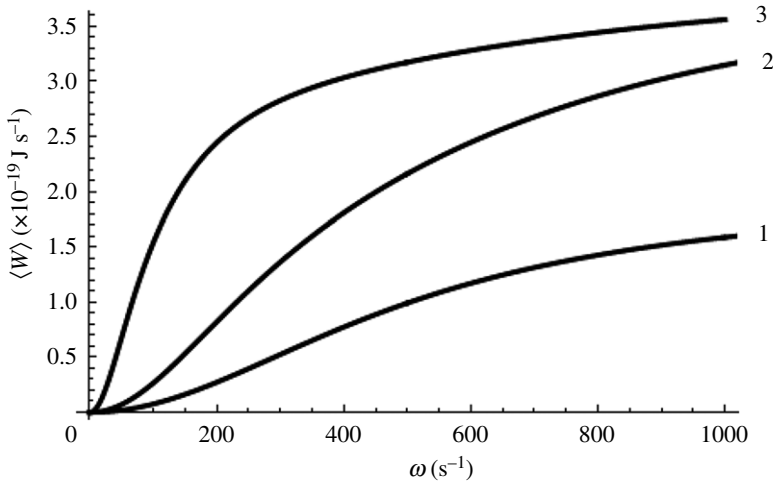


Figure 7. Intensity of heat production $\langle W \rangle$ per particle versus the field frequency ω in the particles with weak magnetic anisotropy. Curve 1, single particle; curve 2, chain of two particles (4.5) and (4.6); curve 3, cluster of three particles in (4.9) and (4.10). The diameter of particles is 10 nm.

Here, r_{ij} and r_{il} are the radius vectors connecting the centres of the corresponding particles; and M_j , M_l and r_{il} are defined as in (3.19).

By using (3.18) and (4.7), one can represent H_i^{eff} as

$$\left. \begin{aligned} H_{x_1} &= H_0 \sin \alpha \cos \omega t + \frac{1}{96} \left(-M_{x_2} - M_{x_3} - 3\sqrt{3}M_{z_2} + 3\sqrt{3}M_{z_3} \right), \\ H_{x_2} &= H_0 \sin \alpha \cos \omega t + \frac{1}{96} \left(8M_{x_3} - M_{x_1} - 3\sqrt{3}M_{z_1} \right), \\ H_{x_3} &= H_0 \sin \alpha \cos \omega t + \frac{1}{96} \left(-M_{x_1} + 8M_{x_2} + 3\sqrt{3}M_{z_1} \right), \\ H_{z_1} &= H_0 \cos \alpha \cos \omega t + \frac{1}{96} \left(-3\sqrt{3}M_{x_2} + 3\sqrt{3}M_{x_3} + 5M_{z_2} + 5M_{z_3} \right), \\ H_{z_2} &= H_0 \cos \alpha \cos \omega t + \frac{1}{96} \left(-3\sqrt{3}M_{x_1} - 4M_{z_3} + 5M_{z_1} \right) \\ \text{and} \quad H_{z_3} &= H_0 \cos \alpha \cos \omega t + \frac{1}{96} \left(3\sqrt{3}M_{x_1} + 5M_{z_1} - 4M_{z_2} \right). \end{aligned} \right\} \quad (4.8)$$

Combining equations (2.1), (4.3) and (4.8), we come to the following form of the Debye equation for the i th particle magnetization:

$$\left. \begin{aligned} \tau \frac{dM_{x_1}}{dt} + M_{x_1} - \frac{\kappa}{96} \left(-M_{x_2} - M_{x_3} - 3\sqrt{3}M_{z_2} + 3\sqrt{3}M_{z_3} \right) &= \kappa H_0 \cos \omega t \sin \alpha, \\ \tau \frac{dM_{x_2}}{dt} + M_{x_2} - \frac{\kappa}{96} \left(8M_{x_3} - M_{x_1} - 3\sqrt{3}M_{z_1} \right) &= \kappa H_0 \cos \omega t \sin \alpha, \\ \tau \frac{dM_{x_3}}{dt} + M_{x_3} - \frac{\kappa}{96} \left(-M_{x_1} + 8M_{x_2} + 3\sqrt{3}M_{z_1} \right) &= \kappa H_0 \cos \omega t \sin \alpha, \\ \tau \frac{dM_{z_1}}{dt} + M_{z_1} - \frac{\kappa}{96} \left(-3\sqrt{3}M_{x_2} + 3\sqrt{3}M_{x_3} + 5M_{z_2} + 5M_{z_3} \right) &= \kappa H_0 \cos \omega t \cos \alpha, \\ \tau \frac{dM_{z_2}}{dt} + M_{z_2} - \frac{\kappa}{96} \left(-3\sqrt{3}M_{x_1} - 4M_{z_3} + 5M_{z_1} \right) &= \kappa H_0 \cos \omega t \cos \alpha \\ \text{and} \quad \tau \frac{dM_{z_3}}{dt} + M_{z_3} - \frac{\kappa}{96} \left(3\sqrt{3}M_{x_1} + 5M_{z_1} - 4M_{z_2} \right) &= \kappa H_0 \cos \omega t \cos \alpha. \end{aligned} \right\} \quad (4.9)$$

Again, changing $\cos \omega t$ to $\exp(i\omega t)$ and representing $M_i = M_i^0 \exp(i\omega t)$, we get a system of algebraic equations with respect to the complex amplitudes M_i^0 . Having found these components, we determine the intensity of MH heat production per one particle as

$$\langle W \rangle = \frac{1}{4\pi} \mu_0 V H_0 \omega \int_0^{2\pi} \left(- \sum_{i=1}^N \left(\frac{(M_{x_i}^0(\alpha))'' \sin \alpha + (M_{z_i}^0(\alpha))'' \cos \alpha}{N} \right) \right) d\alpha, \quad N=3. \quad (4.10)$$

Figure 7 represents the results of calculations of the intensity $\langle W \rangle$ of heat production by the single particle, and a particle in the two-particle cluster (4.5) and (4.6) and in the ‘triangle’ of three particles (4.9) and (4.10) with negligible magnetic anisotropy. Unlike the situation in the case of the particles with strong magnetic anisotropy, clusterization of the particles with weak anisotropy enhances the thermal effect.

5. Conclusion

The results of the mathematical modelling of MH heat production by immobilized single-domain ferromagnetic particles are presented. Our results demonstrate that clusterization of the particles enhances the produced thermal effect in the case of weak (negligible) magnetic anisotropy of the particles, and weakens this effect in the case of strong anisotropy. In the last case, the effect depends on the law of the particles’ axes of easy magnetization in the clusters. We believe that this conclusion must be taken into account in biomedical applications of magnetic hyperthermia.

Data accessibility. This article has no additional data.

Competing interests. We declare we have no competing interests.

Funding. This work has been done with the financial support of a programme of the Ministry of Education and Science of the Russian Federation, projects 02.A03.21.0006 and FEUZ-2020-0051, as well as of the Russian Foundation of Basic Research, projects 18-08-00178, 19-52-45001 and 20-02-00022.

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